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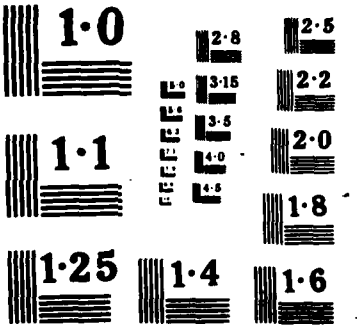
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PICOSECOND NONLINEAR RESONANT INTERACTIONS IN SEMICONDUCTORS

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from

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This research was aimed at advancing understanding and utilization of selected optical properties of semiconductors containing magnetic elements. Emphasis was placed on the interaction of such materials with ultrashort pulses of laser radiation in order to study coupled electronic and magnetic excitations under selected nonequilibrium conditions. We hoped to generate novel results through experimental research for applications to fast optoelectronic devices. The mixed crystal semiconductors (Cd, Mn)Se and (Cd, Mn)Te were used. The contract work has generated a number of "firsts", e.g. we measured the formation of local, microscopic magnetically oriented "domains" through real-time spectroscopy with picosecond laser pulses.

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1. Research Objectives and Summary

The objective of this phase of our research under AFOSR Contract F49620-82 C-0044 (1/1/83 to 12/31/83) was the study of coupled electronic and magnetic excitations in II-VI compound semiconductors containing magnetically active ions with emphasis was placed on the interaction of such materials with ultrashort pulses of laser radiation.The mixed crystal semiconductors (Cd,Mn)Se and (Cd,Mn)Te have provided the material basis for our work. The contract work has been quite successful is generating a 'first' during the investigations of magneto-optical properties of these semiconductors by picosecond laser pulses. In particular with (Cd,Mn)Se we have measured the formation of local, magnetically oriented 'domains', the so-called bound magnetic polaron effects, through real-time spectroscopy. The magnetic polarons here form through the spin exchange interaction of a localized exciton with the Mn-ion magnetic moments within its effective Bohr orbit. Typical formation times at low temperatures (< 10 K) have been measured to be on the order of 400-800 psec.

The research results derived from this AFOSR sponsored research have formed the basis of a number of scientific publications, as enumerated below. In addition to regular scientific meetings, the principal investigator has been invited to present the research results in internationally recognized forums.

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Chief, Technical Information Division

2. Research Accomplishments and Results

Formation of the Bound Magnetic Polaron in (Cd,Mn)Se

In this phase of the AFOSR supported research we have obtained results where for the first time evidence for the formation of a bound magnetic polaron (BMP) is directly obtained in time-resolved experiments by picosecond laser spectroscopy. In particular, we have examined the time evolution of spectra associated with the neutral donor bound exciton in n-$Cd_{0.90}Mn_{0.10}Se$. Our recent results from cw photoluminescence work in $Cd_{1-x}Mn_xSe$ have shown how the BMP contribution to the energy of a neutral donor bound exciton is likely to be significantly larger than that for the neutral donor.[1] This is attributed to the smaller effective Bohr radius of the exciton and the larger exchange constant for the (outer valence) hole, and manifests itself in a large additional low-energy shift of the bound-exciton luminescence peak for x = 0.10 composition as the temperature is lowered from 10 to 2K. Furthermore, a significant apparent linewidth broadening was measured. In the interpretation of Ref. 1 this behavior reflects the transition of the bound exciton-spin complex from the thermodynamic-fluctuation-dominated regime to the bound-polaron regime. In the work reported here, we show evidence for the formation of a BMP associated with the bound exciton and connect the measured formation time to the Mn^{2+}-ion spin-spin relaxation processes.

The experimental arrangement employed to perform time-resolved spectroscopy is based on the use of two synchronously pumped, wavelength-tunable dye lasers in an excite-probe configuration, as detailed elsewhere.[2] In the present instance, a picosecond pulse of excitation promotes a low density of bound excitons (on the order of $1\times10^{14}cm^{-3}$). The presence of this excitation is measured by a weak time-delayed probe pulse as an induced

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change in its transmission [$\Delta T(\omega)/T(\omega)$] through the illuminated sample volume. Under circumstances considered here and for $\Delta T > 0$, the dominant contribution to such optically modulated probe signals is directly proportional to the number of donor sites occupied by the excitons at a give instant of time and energy. This follows by noting that

$$\Delta T/T \sim -\Delta\alpha l \sim \Delta N_{ex} \times f(\omega) l$$

where ΔN_{ex} is the exciton density, $f(\omega)$ a line-shape function, and l the sample thickness.

Figure 1 summarizes the main features in our data at T = 2 K and shows modulated problem transmission spectra recorded at different delays (following the excitation at $t = 0$) at a photon energy of $\hbar\omega_{ex} = 1.968$ eV, somewhat below the free-exciton energy. The spectra have been properly normalized by accounting for the variation in the absorption coefficient, void of any fine structure.[3] By making comparisons with steady-state luminescence spectra,[1] observations from our earlier time-resolved work at higher temperatures, and particularly the appearance of a distinct peak in the *excitation* spectrum at short time delays, we can unambiguously assign the spectral features shown to a bound-exciton complex. The initial formation of a bound exciton for such an excitation energy was confirmed to be rapid ($\lesssim$20 psec) on the time scale of interest here. Most importantly, the data in Fig. 1 show a clear *time-dependent* shift in the probe spectra. In remarkable contrast, we found that the time-resolved spectra observed at 10 K and above (as illustrated in Ref. 3 exhibited *no* measurable shifts but merely a time-dependent amplitude, consistent with the formation and decay of an energetically stationary bound exciton. This constant spectral position of the peak of the modulated transmission at 10 K is shown as the dashed line in Fig. 1. At 2 K we measure an overall red shift in excess of 10 meV within

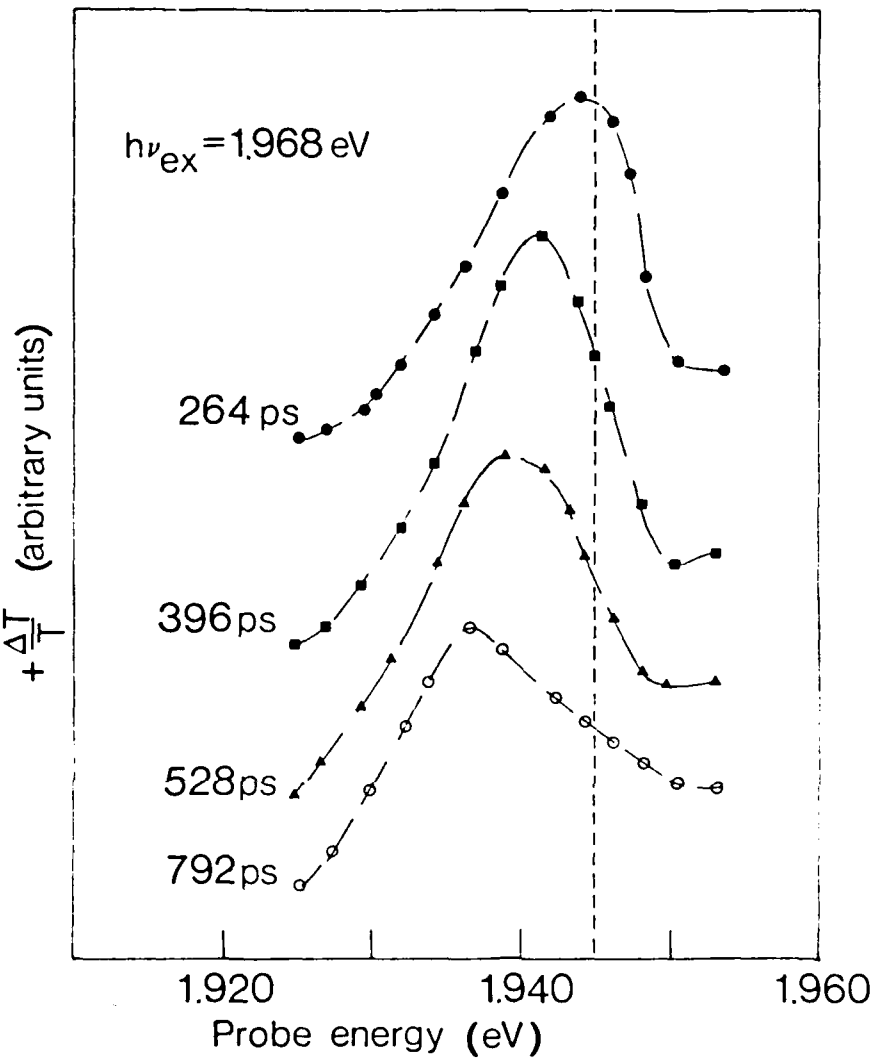

Figure 1: Modulated transimission spectra for the bound exciton in $Cd_{0.90}Mn_{0.10}Se$ at 2K as a function of excite-probe delay in picoseconds (excitation at 1.968 eV). Interpolation of data appears as the dashed line. Relative amplitudes are not fully normalized in this representation. Vertical dashed line denotes the position of the spectrally stationary peak at T= 10K.

the 800-psec range of our experimental setup. Figure 2 shows the position of the probe spectral peaks as a function of the delay. A good fit to the data can be made with a single exponential time constant of 400 psec. At the same time, the measured single-amplitude decay indicates a lifetime for the bound exciton complex of some 600 psec. The latter is longer by approximately a factor of 3 than a similarly measured lifetime at 10 K.

An additional crucial observation is related to the dependence of the probe spectra on the excitation energy. At all temperatures monitored, a resonancelike feature appeared in the excitation spectrum (as shown at 10 K in Ref. 10). At 2 K, this enhancement had an approximate low-energy limit at about 1.945 eV, below which the probe spectra of Fig. 1 decreased precipitously in amplitude. This provides direct evidence that the time-dependent behavior in Fig. 1 involves a dynamic self-energy relaxation of a bound-exciton state *only initially* accessible for excitation. Some small-amplitude probe signals could be seen when exciting at further lower energies but these contributed to much broader spectra which were also temporally distinctly separate from the bound-exciton kinetics. They appear likely to be connected to localization phenomena from alloy potential fluctuations, coupled with other impurity-related effects probably responsible also for the observed absorption-edge broadening.

We believe that the above observations provide strong evidence for the existence of a bound magnetic polaron in a semiconductor from a direct measurement of its dynamics of formation. The time-dependent Stokes shifts evident in Fig. 1 are here interpreted as corresponding to the different stages of the evolution toward spin ordering about the impurity sites. The occurrence of "polaronic relaxation" is also clearly indicated by the experimental ability to couple excitation directly only to the initial "bare" (spin-uncorrelated)

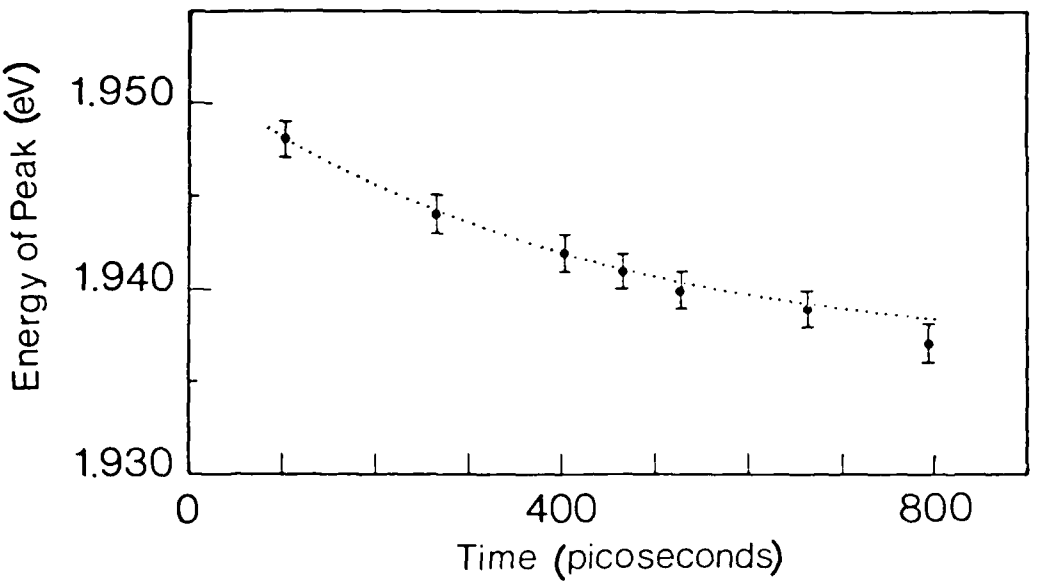

Figure 2: Energetic position of the peak of the exciton line as a function of time delay. Excitation energy in each case is 1.968 eV. Dotted line shows a simple exponential fit.

bound-exciton state. When comparing the steady-state photoluminescence spectra of Huber *et al.*[1] at 2 K with those in Fig. 1, we find good agreement with their results when time integrating our spectra. This explains the puzzle connected with the anomalously broad linewidths reported in Ref. 1. Furthermore, at a higher temperature of 10 K the transient (instantaneous) and steady-state spectra are in mutual agreement while showing frustration of full polaron formation due to thermal disorder.

Dynamically, the BMP formation can be viewed as an evolution of the bound exciton from an initial statistical regime where a thermally fluctuating individual Mn^{2+}-ion spin has not correlation with the charge carrier spin. To first order the carrier here is represented by the outer hole of the bound exciton. (Our experiment is sensitive to such fluctuations since a large number of laser pulses are used to accumulate each data point.) An inhomogeneously broadened optical linewidth, associated with transitions originating from a distribution of perturbed and spin-split levels is then expected form alloy compositional fluctuations.[1] In the transient experiment such an initial state is prepared by the excitation from an applied laser pulse. At 2 K, the exchange interaction between the carrier spin and those of the ion leads to a mutually correlated spin alignment, the BMP, which in the notation of Ref. 5 can be expressed as an effective magnetization of the form $M(r) = \eta\exp(-2r/a_x)$, where η is a variationally determined amplitude and a_x the self-consistent Bohr radius of the carrier (in the hydrogenic envelope approximation). The dynamic evolution of the spectra of Fig. 1 is a direct measurement of the development of such magnetization. In the one-electron (hole) model the mean energy shift E_m associated with the lower spin-split level of the exciton can be calculated form the model of Dietel and Spalek in the BMP (mean-field) limit as[4]:

$$E_M = \beta\chi\tanh(E_M/2kT)/(32\pi g^2\mu_B a_x^3) \quad , \qquad (1)$$

where β is the exchange constant for the outer hole, χ the susceptibility and a_x the Bohr radius. Quantitatively, we find a reasonable agreement of our data with the predicted energy shifts for the formation of the BMP in $Cd_{0.90}Mn_{0.10}Se$. More specifically the BMP contribution adds approximately 15 meV to the exciton binding energy for an assumed Bohr radius of 30 A by using a exchange constant of $-\beta N_o$ = 1.15 eV[5] where N_o is the number of cations per unit volume. Here we assume that the final state associated with the probe laser transition, i.e., the neutral donor, experiences a much smaller energy shift as shown earlier.[1] In the BMP regime, the alloy compositional fluctuations are still expected to contribute to inhomogeneous linewidths which remain rather broad. One potential complication affecting these estimates concerns the possible participation of the inner pair of electrons on the exchange interactions. Conceivably, the large effective internal magnetic fields (well in excess of 100kG) could alter the bound exciton configuration from that assumed by us.

In conclusion, we have obtained strong evidence form direct time-resolved measurements for the evolution of a photoinduced BMP associated with an impurity bound exciton in $Cd_{0.90}Mn_{0.10}Se$. These results can be connected to previously measured cw photoluminescence spectra which provide a time-integrated contrast to our spectra. While reasonable quantitative agreement exists between experiment and estimates for the BMP energy and formation time in a simple one-particle approach, questions remain concerning the exciton configuration in the BMP limit and the contribution to its formation time by collective spin effects.

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3. Scientific Publications and Presentations Resulting from the AFOSR Supported Research

1. "Picosecond Kinetics of the Donor Bound Exciton in (Cd,Mn)Se", J. H. Harris and A. V. Nurmikko, Phys. Rev. B28, 1181 (1983).

2. "Formation of the Bound Magnetic Polaron in (Cd,Mn)Se", J. H. Harris and A. V. Nurmikko, Phys. Rev. Lett. 51, 1472 (1983).

3. "Subnanosecond Luminescence Spectroscopy of (Cd,Mn)Se under High Intensity Optical Excitation", C. A. Huber and A. V. Nurmikko, Solid State Comm. 48, 675 (1983).

4. "Picosecond Modulated Reflectance in Semiconductors", A. V. Nurmikko, in Ultrafast Processes in Semiconductors, R. R. Alfano, Ed., Vol. II, p. 509, Academic Press (1984).

In addition to these scientific publications, the AFOSR supported work has been presented in numerous scientific conferences.

1. Bell Laboratories, Seminar, Holmdel, NJ, April 1983.
2. M.I.T. Magnet Laboratory, Seminar, Cambridge, MA, May 1983.
3. Technical University of Helsinki, Colloquium, Helsinki, Finland, June 1983.
4. Optical Society of America Annual Meeting, New Orleans, October 1983.
5. 8th Int. Conf. of IR and MM-waves, Miami, December 1983.
6. University of Maryland, Physics Department Seminar, December 1983.

4. Personnel

The following have had direct support from this AFOSR grant:

Professor A. V. Nurmikko, Principal Investigator;

Carmen A. Huber, Ph.D. candidate.

5. Patents

No patents have been filed in connection with this AFOSR sponsored research.

6. *Remaining Funds*

No remaining funds exist.

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